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(54) Title: FLAME RETARDANT SYNDIOTACTIC POLYSTYRENE RESIN COMPOSITION

(57) Abstract: The syndiotactic polystyrene resin composition according to the present invention comprises (a) 100 parts by weight of a syndiotactic polystyrene, (b) about 1 ~ 50 parts by weight of a hydrogenated styrene/butadiene/styrene block copolymer (SEBS) as a rubber component to improve impact strength of the resin composition, (c) about 1 ~ 15 parts by weight of a hydrogenated styrene/butadiene/styrene - graft-maleic anhydride (SEBS-MA) as a first compatibilizer to improve compatibility between the syndiotactic polystyrene and the rubber, (d) about 1 ~ 90 parts by weight of polyphenyleneoxide (PPO) as a second compatibilizer to improve compatibility between the syndiotactic polystyrene and the rubber, and (e) about 1 ~ 60 parts by weight of a phosphoric acid ester as a halogen-free flame retardant. The syndiotactic polystyrene resin composition may optionally include (f) about 1 ~ 70 parts by weight of glass fibers.

Flame Retardant Syndiotactic Polystyrene Resin Composition

Field of the Invention

The present invention relates to a syndiotactic polystyrene resin composition using a halogen-free flame retardant, which provides good flame retardancy as well as sufficient other physical properties. More particularly, the present invention relates to a syndiotactic polystyrene resin composition that can show good impact strength by containing a rubber component and good flame retardancy by using a halogen-free flame retardant.

Background of the Invention

Syndiotactic polystyrene is a very attractive polymer. The polymer shows low specific gravity, low dielectric constant, high flexural modulus, good heat resistance, good rigidity, high melting point (T_m) of about 270℃ and excellent resistance to chemicals. Accordingly the syndiotactic polystyrene has become a promising material for various applications in the automotive, electronic and packaging industries. However, the applications of the syndiotactic polystyrene resin composition are severely limited because impact strength is too low. To overcome such shortcoming, an impact modifying material such as rubber and various compatibility agents for the syndiotactic polystyrene and the impact modifying material have been used in the syndiotactic polystyrene resin composition.

U.S. Patent No. 5,270,353 to Nakano et al. discloses a styrene-based resin composition comprising a styrene-based polymer having a syndiotactic configuration and no functional group, a styrene-based polymer having an epoxy group and an inorganic filler surface treated by a silane compound or a titanium compound.

U.S. Patent No. 5,352,727 to Okada discloses a polystyrene composition comprising syndiotactic polystyrene and a syndiotactic polystyrene impact modifier,

said syndiotactic polystyrene impact modifier being the reaction product of a syndiotactic polystyrene having a first reactive functional group, and a rubbery elastomer containing a second reactive functional group, said first and second reactive functional groups being reactive with each other.

U.S. Patent No. 5,391,603 to Wessel et al. discloses a composition comprising a syndiotactic, vinyl aromatic thermopalstic resin, a rubbery, impact absorbing domain forming polymer, a nucleator for the syndiotactic, vinyl aromatic thermoplastic resin, a mineral, glass, ceramic, polymeric or carbon reinforcing agent, and one or more polyarylene ethers or polar group modified polyarylene ethers.

U.S. Patent No. 5,543,462 to Okada et al. discloses an impact resistant polystyrene composition comprising a styrenic polymer having syndiotactic configuration and a rubbery elastomer modified by a modifier having a polar group, and optionally a rubbery elastomer and/or a poly(phenylene ether).

U.S. Patent No. 5,654,365 to Havriliak, Jr. et al. discloses a syndiotactic polystyrene composition impact-modified by a blend of polystyrene- hydrogenated polybutadiene block copolymers and one or more core/shell modifiers based on a lightly or non-crosslinked rubber core.

U.S. Patent No. 5,760,105 to Okada et al. discloses a styrenic resin composition comprising a styrenic polymer having syndiotactic configuration (sPS) or modified sPS, a thermoplastic resin having a polar group, a rubbery elastomer having affinity for the sPS and a compatibilizer having a polar group.

U.S. Patent No. 5,777,028 to Okada et al. discloses an impact resistant polystyrene composition comprising a styrenic polymer having syndiotactic configuration, a rubbery elastomer having an olefinic component or a polyolefin and a styrene/olefin block or graft copolymer having a microphase separation temperature of 180 ℃.

Idemitsu Co. of Japan had sold a syndiotactic polystyrene resin composition which is reinforced with glass fibers to improve impact strength and employs a halogen-containing flame retardant.

There are two flame retardants for use of a resin composition, a halogen-containing flame retardant and a halogen-free flame retardant. The halogen-

containing flame retardant includes bromides and chlorides and the halogen-free flame retardant includes phosphoric acid esters.

Resin compositions containing a halogen-containing flame retardant have good flame retardancy and resistance to chemicals. However, the resin compositions have poor resistance to weather, become poor in heat stability during molding process at high temperature to generate halogenated gases resulting erosion of the mold, and generate toxic gases during combustion to cause an environmental problem. On the other hand, resin compositions containing a phosphoric acid ester of the halogen-free flame retardant form a great deal of char due to carbonization, dehydration and additional plasticization.

The present inventors has filed two Korean patent applications (Korean Application Nos. 1999-39858 and 1999-39859), which relate to syndiotactic polystyrene compositions having an improved flame retardancy as well as maintaining mechanical strength and impact strength.

The present inventors have developed a new syndiotactic polystyrene resin composition which comprises (a) a syndiotactic polystyrene, (b) a hydrogenated styrene/butadiene/styrene block copolymer (SEBS) as a rubber component to improve impact strength of the resin composition, (c) a hydrogenated styrene/butadiene/styrene - graft - maleic anhydride (SEBS-MA) as a first compatibilizer to improve compatibility between the syndiotactic polystyrene and the rubber, (d) polyphenyleneoxide (PPO) as a second compatibilizer to improve compatibility between the syndiotactic polystyrene and the rubber, and (e) a phosphoric acid ester as a halogen-free flame retardant.

Objects of the Invention

A feature of the present invention is the provision of a syndiotactic polystyrene resin composition which is not harmful to human bodies and does not cause a n environmental problem by employing phosphoric acid ester as a halogen-free flame retardant.

Another feature of the present invention is the provision of a syndiotactic

polystyrene resin composition having an improved impact strength by containing a hydrogenated styrene/butadiene/styrene block copolymer (SEBS) as a rubber component.

A further feature of the present invention is the provision of a syndiotactic polystyrene resin composition having good physical properties which are required for various applications thereof.

Other objects and advantages of this invention will be apparent from the ensuing disclosure and appended claims.

Summary of the Invention

The syndiotactic polystyrene resin composition according to the present invention comprises (a) 100 parts by weight of a syndiotactic polystyrene, (b) about 1 ~ 50 parts by weight of a hydrogenated styrene/butadiene/styrene block copolymer (SEBS) as a rubber component to improve impact strength of the resin composition, (c) about 1 ~ 15 parts by weight of a hydrogenated styrene/butadiene/styrene - graft - maleic anhydride (SEBS-MA) as a first compatibilizer to improve compatibility between the syndiotactic polystyrene and the rubber, (d) about 1 ~ 90 parts by weight of polyphenyleneoxide (PPO) as a second compatibilizer to improve compatibility between the syndiotactic polystyrene and the rubber, and (e) about 1 ~ 60 parts by weight of a phosphoric acid ester as a halogen-free flame retardant. The syndiotactic polystyrene resin composition may optionally include (f) about 1 ~ 70 parts by weight of glass fibers.

Detailed Description of the Invention

The syndiotactic polystyrene resin composition according to the present invention comprises (a) a syndiotactic polystyrene, (b) a hydrogenated styrene/butadiene/styrene block copolymer (SEBS) as a rubber component, (c) a hydrogenated styrene/butadiene/styrene - graft - maleic anhydride (SEBS-MA) as a first compatibilizer, (d) polyphenyleneoxide (PPO) as a second compatibilizer, and

(e) a phosphoric acid ester as a halogen-free flame retardant. All the components will be described in detail as follow:

(A) Syndiotactic Polystyrene (sPS)

In general, polystrenes are classified into an atactic, an isotactic and a syndiotactic structure depending on the position of benzene rings as side chains. An atactic polystyrene has an irregular arrangement of the benzene rings and an isotactic polystyrene has an arrangement that the benzene rings are positioned at one side of the polymer main chain. On the other hand, a syndiotactic polystyrene has a regularly alternating arrangement of the benzene rings.

The syndiotactic polystyrenes are prepared with styrene monomers under the catalyst system a metallocene catalyst and a cocatalyst. The metallocene catalyst has a bridged structure of a complex of Group IV transition metals such as Ti, Zr, Hf etc. in the Periodic Table and a ligand having one or two cycloalkane dienyl groups such as cyclopentadienyl group, indenyl groups, fluorenyl groups, and derivatives thereof. As a metallocene catalyst has a high activity, the metallocene catalyst can prepare a polymer having better physical properties than the Ziegler-Natta catalyst. U.S. Patent Serial Nos. 08/844,109 and 08/844,110 disclose new alkyl-bridged binuclear metallocene catalysts, silyl-bridged binuclear metallocene catalysts, and alkyl-silyl-bridged binuclear metallocene catalysts to polymerize styrene monomer to produce polystyrene having a high stereoregularity, a high melting point, and a good molecular weight distribution.

U.S. Patent Serial No. 09/231,884 filed January 14, 1999 discloses a new syndiotactic polystyrene prepared under the catalyst system consisting of a metallocene catalyst and a cocatalyst.

It is preferable to use a syndiotactic polystyrene with a syndiotacticity of 97 % or more in this invention.

(B) Hydrogenated Styrene/Butadiene/Styrene Block Copolymer (SEBS)

A rubber component is added to the syndiotactic polystyrene resin composition according to the present invention. A hydrogenated styrene/butadiene/styrene block copolymer (SEBS) is preferably used as a rubber component. The rubber component absorbs impact and improves impact strength of the resin composition.

The hydrogenated styrene/butadiene/styrene block copolymer (SEBS) is used in the amount of about 1 ~ 50 parts by weight per 100 parts of the syndiotactic polystyrene.

(C) Hydrogenated Styrene/Butadiene/Styrene - graft - maleic anhydride (SEBS-MA)

A hydrogenated styrene/butadiene/styrene – graft - maleic anhydride (SEBS-MA) is used in the syndiotactic polystyrene resin composition according to the present invention as a first compatibilizer to improve compatibility between the syndiotactic polystyrene and the rubber component.

The hydrogenated styrene/butadiene/styrene – graft - maleic anhydride (SEBS-MA) is prepared by treating hydrogenated styrene/butadiene/styrene with maleic acid anhydride, which can be carried out by an ordinary skilled person in the art. Also, the hydrogenated styrene/butadiene/styrene – graft - maleic anhydride (SEBS-MA) is commercially available.

The hydrogenated styrene/butadiene/styrene – graft - maleic anhydride (SEBS-MA) is used in the amount of about 1 ~ 15 parts by weight per 100 parts of the syndiotactic polystyrene.

(D) Polyphenyleneoxide (PPO)

A second compatibilizer is used in the syndiotactic polystyrene resin composition according to the present invention to improve compatibility between the syndiotactic polystyrene and the rubber component. Polyphenyleneoxide (PPO) is preferably used as a second compatibilizer in this invention. Polyphenyleneoxide (PPO) is commercially available.

Polyphenyleneoxide (PPO) is used in the amount of about 1 ~ 90 parts by weight per 100 parts of the syndiotactic polystyrene.

(E) Phosphoric Acid Ester

A phosphoric acid ester is used in the syndiotactic polystyrene resin composition according to the present invention to provide the resin composition with flame retardancy. The phosphoric acid ester is a halogen-free flame retardant. Resorcinol bis(di-2,6-xylenyl phosphate) can be preferably used. The resin composition containing a phosphoric acid ester of the halogen-free flame retardant forms a great deal of char due to carbonization, dehydration and additional plasticization to provide an improved flame retardancy. The halogen-free flame retardant does neither generate toxic gases which are harmful to human bodies, nor cause an environmental problem.

The phosphoric acid ester is used in the amount of about 1 ~ 60 parts by weight per 100 parts of the syndiotactic polystyrene.

The syndiotactic polystyrene resin composition according to the present invention may optionally include a reinforcing material.

(F) Glass Fibers

A reinforcing material may be added to the syndiotactic polystyrene resin composition according to the present invention. Glass fibers may be preferably used as a reinforcing material in this invention.

Glass fibers are used in the amount of about 1 ~ 70 parts by weight per 100 parts of the syndiotactic polystyrene.

Other additives may be added to the resin composition of the present invention, when necessary. The additives include an additional flame retardant, a lubricant, a releasing agent, an anti-dripping agent, an impact modifier, a plasticizer,

a heat stabilizer, an anti-oxidant, a light stabilizer, pigments, dye and the like. An inorganic filler such as talc, silica, mica or ceramic can be added too. The additives are employed in an amount of about 0 to 40 parts by weight as per 100 parts of the syndiotactic polystyrene.

The invention may be better understood by reference to the following examples which are intended for the purpose of illustration and are not to be construed as in any way limiting the scope of the present invention, which is defined in the claims appended hereto. In the following examples, all parts and percentage are by weight unless otherwise indicated.

Examples

The components to prepare the syndiotactic polystyrene resin compositions of Examples 1-3 and Comparative Examples 1-2 are as follows:

(A) Syndiotactic Polystyrene

Syndiotactic polystyrene with syndiotacticity of 97 % was prepared in a glass reactor.

(B) Hydrogenated Styrene/Butadiene/Styrene Block Copolymer (SEBS)

A commercial product (Kraton G-1651) of Shell Co. was used as SEBS.

(C) Hydrogenated Styrene/Butadiene/Styrene – graft-maleic anhydride (SEBS-MA)

A commercial product (M-1913) of Asahi Co. of Japan was used as SEBS-MA.

(D) Polyphenyleneoxide (PPO)

A commercial product of Asahi Co. of Japan was used as PPO.

(E) Phosphoric Acid Ester

A commercial product (PX-200) of Daihachi Chemical Co. of Japan was used as a phosphoric acid ester in Examples 1-2 and Comparative Examples 1-2. Triphenyl phosphate (TPP) of Daihachi Chemical Co. of Japan was used as a phosphoric acid ester in Example 3.

The components of Examples 1-3 and Comparative Examples 1-2 are shown in Table 1.

Table 1

(parts by weight)

	Examples			Comp. Examples	
	1	2	3	1	2
(A) Syndiotactic Polystyrene	100	100	100	100	100
(B) SEBS	21.0	21.1	21.1	20.9	21.0
(C) SEBS-MA	5.4	5.3	5.3	5.3	5.3
(D) PPO	71.1	88.2	88.2	5.2	38.1
(E) Phosphoric Acid Ester	26.4[1)]	52.7[1)]	52.7[2)]	13.2[1)]	19.7[1)]
Flame Retardancy	V-1	V-0	V-0	fail	fail

Notes: 1, 2) PX-200 and Triphenyl phosphate (TPP) is made by Daihachi Chemical Co. of Japan

For testing flame retardancy of the resin compositions of Examples 1-3 and Comparative Examples 1-2, specimens of 1/8", 1/16" and 1/32" were prepared and tested in accordance with UL 94. The specimens of Example 1 showed flame retardancy of V-1, and the specimens of Examples 2-3 showed flame retardancy of

V-0. The test results were shown in Table 1.

For measuring physical properties of the resin compositions of Examples 1-3, the resin compositions were prepared in pellets in a twin screw extruder at 280 ℃. The pellets were dried at 80 ℃ for 3 hours and molded into specimens in a 50 ton injection-molder at 300 ℃. The physical properties such as yield strength, breaking strength, elongation, flexural strength and Izod impact strength were measured and shown in Table 2.

Table 2

	Examples		
	1	2	3
yield strength (kgf/cm^2)	647	607	407
breaking strength (kgf/cm^2)	414	466	265
elongation (%)	28	40	45
flexural strength (kgf/cm^2)	26,300	26,160	19,380
Izod impact strength (kgf/cm^2)	32	24	33

Note: The yield strength, breaking strength and the elongation were measured in accordance with ASTM D638 and flexural strength were measured in accordance with ASTM D790 and Izod impact strength was measured in notched type in accordance with ASTM D256.

As shown in Table 2, when the flame retardancy improves, the physical properties improve. When the amount of rubber component decreases, the impact strength becomes lower. But the more PPO is employed, that is a function of

compatible agent between syndiotactic polystyrene and rubbery phase Therefore, the resin composition according to the present invention has a good flame retardancy and is applicable to articles in which a certain impact strength is not required.

The present invention can be easily carried out by an ordinary skilled person in the art. Many modifications and changes may be deemed to be with the scope of the present invention as defined in the following claims.

What is claimed is:

1. A syndiotactic polystyrene resin composition comprising (a) 100 parts by weight of a syndiotactic polystyrene, (b) about 1 ~ 50 parts by weight of a hydrogenated styrene/butadiene/styrene block copolymer (SEBS) as a rubber component to improve impact strength of the resin composition, (c) about 1 ~ 15 parts by weight of a hydrogenated styrene/butadiene/styrene – graft-maleic anhydride (SEBS-MA) as a first compatibilizer to improve compatibility between the syndiotactic polystyrene and the rubber, (d) about 1 ~ 90 parts by weight of polyphenyleneoxide (PPO) as a second compatibilizer to improve compatibility between the syndiotactic polystyrene and the rubber, and (e) about 1 ~ 60 parts by weight of a phosphoric acid ester as a halogen-free flame retardant.

2. The syndiotactic polystyrene resin composition as defined in claim 1, further comprising about 1 ~ 70 parts by weight of glass fibers per 100 parts of the syndiotactic polystyrene (a).

3. The syndiotactic polystyrene resin composition as defined in claim 1, further comprising an additional flame retardant, a lubricant, a releasing agent, an anti-dripping agent, an impact modifier, a plasticizer, a heat stabilizer, an anti-oxidant, a light stabilizer, pigments, dye and/or an inorganic filler.

4. The syndiotactic polystyrene resin composition as defined in claim 3, wherein said inorganic filler is selected from the group consisting of talc, silica, mica and ceramic.

INTERNATIONAL SEARCH REPORT

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A. CLASSIFICATION OF SUBJECT MATTER

IPC7 C08L 25/06, C08L 51/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimun documentation searched (classification system followed by classification symbols)

IPC7 C08L 25/06, C08L 51/04

Documentation searched other than minimun documentation to the extent that such documents are included in the fileds searched

Korean Patents and Applications for Inventions since 1975
Korean Utility Models and Applications for Utility Models since 1975

Electronic data base consulted during the intertnational search (name of data basc and, where practicable, search trerms used)

NPS, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X, E	KR 2001-27880 A (Samsung General Chemicals Co., Ltd) 04 April 2001 see the whole document	1-4
Y	KR 2001-8688 A (Samsung General Chemicals Co., Ltd) 05 February 2001 see the whole document	1-4
Y	JP 07-90157 A (JAPAN SYNTHETIC RUBBER Co. Ltd) 04 April 1995 see the whole document	1-4

☐ Further documents are listed in the continuation of Box C.

☐ See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevence
"E" earlier application or patent but published on or after the international filing date
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"X" document of particular relevence; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
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"&" document member of the same patent family

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